High-Frequency Surface Impedance of Dirty Type-II Superconductors in the Surface-Sheath Regime

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Surface-resistance measurements in the superconducting surface-sheath regime of Pb–In and Pb–Bi alloys are analyzed in the light of the recent theories of dirty type-II superconductors. From the changes of slope of the surface resistance $R(H)$ at H_{c3}, the temperature dependence of the κ_2 parameter can be deduced. For the alloys investigated, it is found that κ_2 varies more strongly with temperature than expected from the dirty-limit theory. It is believed that this reflects strong-coupling effects. From the value of $R(H)$ at H_{c2}, the absolute value of the sheath thickness can also be derived. This is found to agree well, near T_c, with the theoretical predictions of Fink and Kessinger. The analysis of the experimental data suggests that weak-coupling formalism remains applicable in the strong-coupling limit, and that deviations appear as modified temperature dependences of the critical fields. The $H_{c2}(t)$ and $H_{c3}(t)$ found for the two alloys are nevertheless rather close to the weak-coupling predictions.

I. INTRODUCTION

SURFACE-RESISTANCE measurements have proven useful for studies of surface nucleation in superconductivity.[1] The onset of superconductivity at the surface, in a decreasing magnetic field oriented parallel to the surface, is seen as a linear decrease of the surface resistance.[2] Up to the present no direct analysis of the surface resistance in terms of the microscopic theory of superconductivity has been made, even though there is now a well-developed theory for dirty superconductors.[3]

Our purpose is to analyze experimental data obtained with Pb–In and Pb–Bi alloy samples in the light of recent theories. In this first paper we confine ourselves to the analysis of data referring to the geometry $\mathbf{E}_\omega \parallel \mathbf{H}$, where microwave electric field $\mathbf{E}_\omega$ and static magnetic field $\mathbf{H}$ are parallel. This choice is made because in the other geometry, i.e., $\mathbf{E}_\omega \perp \mathbf{H}$ though still with $\mathbf{H}$ parallel to the sample surface, the microwaves can excite collective modes of the order parameter in the sheath regime,[4,5] and these give rise to additional contributions to the surface resistance. Such additional resistance has been observed both in type-I[6,7] and type-II[8] superconductors, but no really satisfactory explanation[7] has so far been given for this extra absorption. A detailed study of this effect is left for the future.[9]

Surface resistance in the sheath regime and $\mathbf{E}_\omega \parallel \mathbf{H}$ geometry has been studied by Rothwarf *et al.*[2] These authors obtained a reasonably good fit to their experimental $R(H)$ curve with a theoretical model which assumes a uniform order parameter in a sheath of thickness $\alpha(H)\xi(t)$ and a single Ginzburg-Landau (GL) parameter $\kappa(t)$. $\xi(t)$ is the temperature-dependent coherence length and t the reduced temperature T/T_c. For $\alpha(H)$, Rothwarf *et al.* take the values calculated by Fink and Kessinger[10] from the GL equations and for $\kappa(t)$ they choose the parameter $\kappa_1(t)$ derived by Maki[3] in the weak-coupling dirty limit. While the phenomenological theory of Rothwarf *et al.* explains the observed $R(H, T)$ reasonably well, one should like to push theory to the point where it will be possible to derive such parameters as $\kappa_1(t)$, $\kappa_2(t)$, and the sheath thickness from measured $R(H, T)$ curves. The present paper is essentially an attempt in this direction.

The outline of the paper is as follows: In Sec. II we give a description of the premises and results of the theory, but reserve details of the surface-impedance derivation to the Appendix. Section III is devoted to experimental aspects and Sec. IV to experimental results and their analysis, with discussion and conclusions given in Sec. V.

II. PRESENTATION OF THE THEORY

In the Appendix we derive an expression for the surface impedance in terms of the position-dependent complex conductivity $Q(\mathbf{r}, \omega)$ as it obtains in the presence of a magnetic field $\mathbf{H} \parallel \mathbf{E}_\omega$.[11] In particular, we are interested in the surface impedance for $H=H_{c3}$ and $H=H_{c2}$, where we can calculate $Q(\mathbf{r}, \omega)$ from the microscopic theory. In the geometry $\mathbf{E}_\omega \parallel \mathbf{H}$, Fig. 1 shows

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[1] D. Saint-James and P. G. de Gennes, Phys. Letters **7**, 306 (1963).

[2] A. Rothwarf, J. I. Gittleman, and B. Rosenblum, Phys. Rev. **155**, 370 (1967).

[3] K. Maki, Physics **1**, 21 (1964); and P. G. de Gennes, Physik Kondensierten Materie **3**, 79 (1964).

[4] H. J. Fink and R. D. Kessinger, Phys. Letters **25A**, 357 (1967).

[5] K. Maki, Progr. Theoret. Phys. (Kyoto) **39**, 1165 (1968).

[6] B. Rosenblum and M. Cardona, Phys. Letters **9**, 220 (1964).

[7] G. Fischer and R. Klein, Phys. Rev. **165**, 578 (1968).

[8] M. Cardona and B. Rosenblum, Phys. Letters **8**, 308 (1964).

[9] K. Maki and G. Fischer, Phys. Rev. (to be published).

[10] H. J. Fink and R. D. Kessinger, Phys. Rev. **140**, A1937 (1965).

[11] K. Maki, in *Treatise on Superconductivity*, edited by R. D. Parks (Marcel Dekker, Inc., New York, to be published).

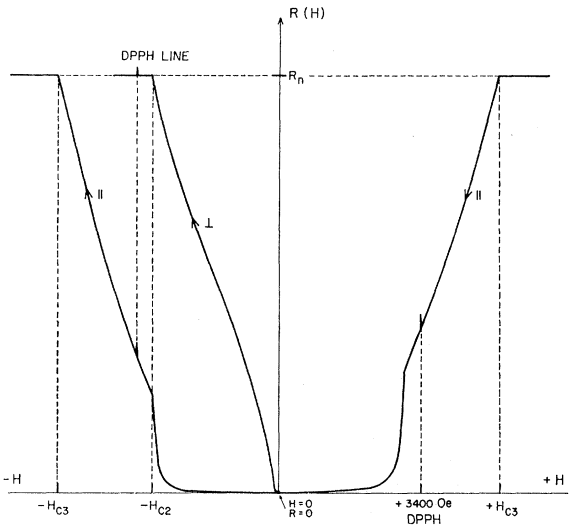

FIG. 1. Recording of $R(H)$ at 4.19°K for a $Pb_{0.91}Bi_{0.09}$ alloy. The diphenyl-picryl-hydrazyl line at 3400 Oe is for field-calibration purposes. The signs || and ⊥ refer to relative orientation of dc magnetic field **H** and sample surface. In the || orientation, **H** is also parallel to the microwave electric field $\mathbf{E}_\omega$.

that $\partial R(H)/\partial H$ has discontinuities at both H_{c3} and H_{c2}. The jump at H_{c3} reflects the nucleation of surface superconductivity, and in the Appendix we derive a theoretical expression for the jump height s_3,

$$s_3=(H_{c3}/R_n)[\partial R(H)/\partial H]|_{H=Hc3}$$
$$=(2\pi)^{1/2}[\delta_0/\xi(t)][2\kappa_2^2(t)-0.334]^{-1}, \quad (1)$$

where $\delta_0=(c^2/2\pi\omega\sigma)^{1/2}$ is the classical normal-state skin depth, σ the dc conductivity, $\omega/2\pi$ the frequency, and $\xi(t)=[\hbar c/2eH_{c2}(t)]^{1/2}$.

The jump at H_{c2} occurs because of the onset of the Abrikosov structure in the bulk of the specimen. Here we find for the jump height s_2,

$$s_2=\frac{H_{c2}}{R_n}\left(\frac{\partial R(H)}{\partial H}\right)\Bigg|_{H=Hc2-0}-\frac{H_{c2}}{R_n}\left(\frac{\partial R(H)}{\partial H}\right)\Bigg|_{H=Hc2+0}$$
$$=r[1-r+r(2/r-1)^{1/2}]\left(\frac{\delta_0}{\xi(t)}\right)^2\{2\beta_A[2\kappa_2^2(t)-1]\}^{-1}, \quad (2)$$

where $r=R(H_{c2})/R_n$ and $\beta_A=1.16$. In the calculation leading to Eq. (2) we have made two important assumptions.

(1) We have assumed that the onset of Abrikosov's structure in the bulk does not modify the surface-sheath structure strongly. This assumption is well supported by the tunneling characteristics of Guyon,[12] which reveal no discontinuity of differential conductance at H_{c2}.

[12] E. Guyon, Advan. Phys. **15**, 417 (1966); see in particular Fig. IV.

(2) The Abrikosov structure is taken to extend right up to the surface and is simply superposed additively to the sheath structure. This is probably the most questionable assumption we are making, as one would expect some transitional region from sheath to Abrikosov structure near the surface, as suggested by Fink.[13]

Equations (1) and (2) unambiguously give the slope discontinuities to be expected in $R(H)$ for a weak-coupling dirty superconductor. However, our alloys show many of the attributes typical of strong-coupling effects, because of the high Pb concentration. Therefore we reverse Eqs. (1) and (2) to derive $\kappa_2(t)$ from the experimental $s_3(t)$ and $s_2(t)$. Our implicit assumption here is that deviations from the weak-coupling dirty superconductors appear simply as a modification of $\kappa_2(t)$. Our data and analysis, as we shall see, strongly suggest that the microscopic weak-coupling formalism remains applicable in the strong-coupling limit, and that deviations appear essentially in the form of slightly modified temperature dependences. In particular, $H_{c2}(t)$ and $H_{c3}(t)$ for our alloys are close to the predictions of the weak-coupling theory[3,14]:

$$\ln(T/T_c)+\psi[\tfrac{1}{2}+(\epsilon_0/4\pi k_B T_c)]-\psi(\tfrac{1}{2})=0, \quad (3)$$

where $\epsilon_0=2DeH_{c2}(t)/\hbar c$, $D=\frac{1}{3}v_F l$ is the electronic diffusion coefficient with l the electron mean free path, and $\psi(z)$ is the digamma function. But the temperature dependence of $\kappa_2(t)$ deduced from $s_3(t)$ is stronger than that deduced from tunneling experiments on In-Bi alloys,[15] reflecting, as we think, the stronger coupling. Analysis of the $s_2(t)$ data, however, leads to $\kappa_2(t)$ values too large by a factor of about 2. We believe that this discrepancy indicates that below H_{c2} our assumption (2) is incorrect. It turns out, in fact, that if we were to introduce a transition region near the surface, over which the Abrikosov structure builds up to its full amplitude in the interior, agreement could be obtained. At this stage we could do this only in an arbitrary phenomenological way, since we have no microscopic theory of the structure of the pair potential near the surface just below H_{c2}.

Whereas Eq. (2) is dependent on the two above assumptions, it is not dependent on the form of the sheath structure itself. However, our experiments give evidence that the square of the pair potential in the range from H_{c3} to below H_{c2} varies almost linearly throughout, i.e., like $(H_{c3}-H)$, as seen in Eq. (A 14). If we express the nonlinearity by means of a factor $\alpha(H,t)$, then $\alpha(H,t)$ can be looked upon as the ratio of the sheath thickness at a field H to the sheath thickness at H_{c3}. Our theory then gives a formula enabling us to calculate $\alpha(H,t)$ in terms of the experimental $R(H)$

[13] H. J. Fink, Phys. Rev. Letters **14**, 853 (1965).
[14] E. Helfand and N. R. Werthamer, Phys. Rev. **147**, 288 (1966).
[15] E. Guyon, F. Meunier, and R. S. Thompson, Phys. Rev. **156**, 452 (1967).

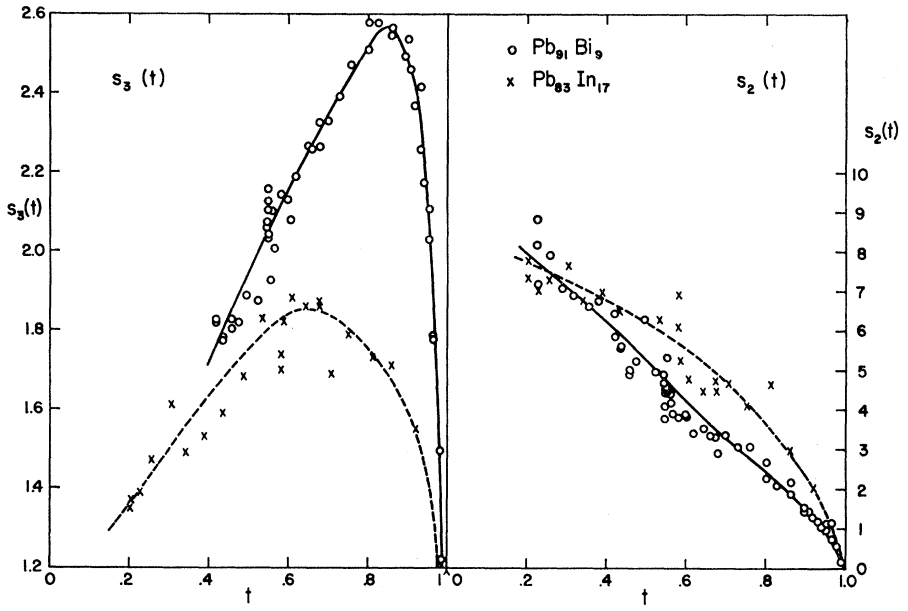

FIG. 2. The experimental functions $s_2(t)$ and $s_3(t)$ for the two alloys $Pb_{0.91}Bi_{0.09}$ and $Pb_{0.83}In_{0.17}$.

curves; specifically, at H_{c2} we obtain

$$\alpha(t)=\alpha(H_{c2},t)=\frac{(2/r-1)^{1/2}-1}{s_3(t)}\left(\frac{H_{c3}}{H_{c3}-H_{c2}}\right). \quad (4)$$

Our data lead to values of $\alpha(t)$ in very close agreement with the theory of Fink and Kessinger.[10] This theory is truly valid only near T_c and our data indicate that below T_c the values of $\alpha(t)$ deviate in a systematic way from $\alpha(1)$.

III. EXPERIMENTAL ASPECTS

We are experimenting at a fixed frequency of 9.5 GHz and our method of measuring the surface resistance $R(H)$ has been abundantly described before.[16] We shall, therefore, deal here only with the question of sample preparation. The Pb–In alloys were found to be very ductile and easy to prepare, following the method of Rothwarf *et al.*[2] Alloys with up to 70% In crystallize in the Pb structure[17] and their critical temperatures decrease by only 10% over the entire range.[18] This suggests that the density of states at the Fermi surface, $N(0)$, does not undergo any abrupt changes when this large amount of a tervalent element like In is alloyed to Pb. When Pb is alloyed with Bi, a quite different behavior is observed. The alloys are rather hard and we did not succeed in producing suitable samples with smooth surfaces by simply pressing the alloys between highly polished metal blocks as had been done with the In alloys. After many attempts the following procedure was found to give very satisfactory samples which yielded sharply featured $R(H)$ curves.

TABLE I. Collected data of the present analysis.

Substance	Pb	$Pb_{0.91}Bi_{0.09}$	$Pb_{0.83}In_{0.17}$
$1/\sigma$ ($\mu\Omega$ cm)	•••	8.5	12
δ_0 (μ)	•••	1.50	1.79
T_c (°K)	7.20	7.65	7.10
H_{c2} (Oe)	594[a]	4870	5090
$\xi(0)$ (Å)	800	260	254
$v_F l$ (cm²/sec)	•••	36	32
$N(0)$ (10^{34}/erg cm³)	1.67[b]	1.92	1.53
H_0 (Oe)	802.6	915	759
$-h_0'(1)$	2.13[c]	1.94	1.63
$\kappa(1)$	0.32_8[a]	2.80	4.20
$\kappa_1(0)$	0.52_3[a]	3.76	4.74
$\kappa_1(0)/\kappa(1)$	1.59_5[a]	1.34	1.13

[a] Reference 27.
[b] From Eq. (10).
[c] Reference 21.

The alloys are first made by melting together high-purity elements in appropriate proportions. Small amounts of the alloys are evaporated on the sample holders. The samples are then lightly polished on a soft cloth wetted with ethanol and 500-Å Linde B powder. The polishing powder is rinsed off with petroleum ether and, after quickly drying, the samples are sealed in evacuated ampuls and annealed for 48 h at about 50°C below their melting point.

If we consider that when alloying with Bi the Pb structure ceases to be stable at about 18% Bi,[17] that T_c has then risen to about 8°K,[19] and that alloying with

[16] G. Fischer and R. Klein, Physik Kondensierten Materie **7**, 12 (1968).
[17] M. Hansen, *Constitution of Binary Alloys* (McGraw-Hill Book Co., New York, 1958).
[18] J. G. Adler, J. E. Jackson, and T. A. Will, Phys. Letters **24A**, 407 (1967).
[19] J. G. Adler and S. C. Ng, Can. J. Phys. **43**, 594 (1965).

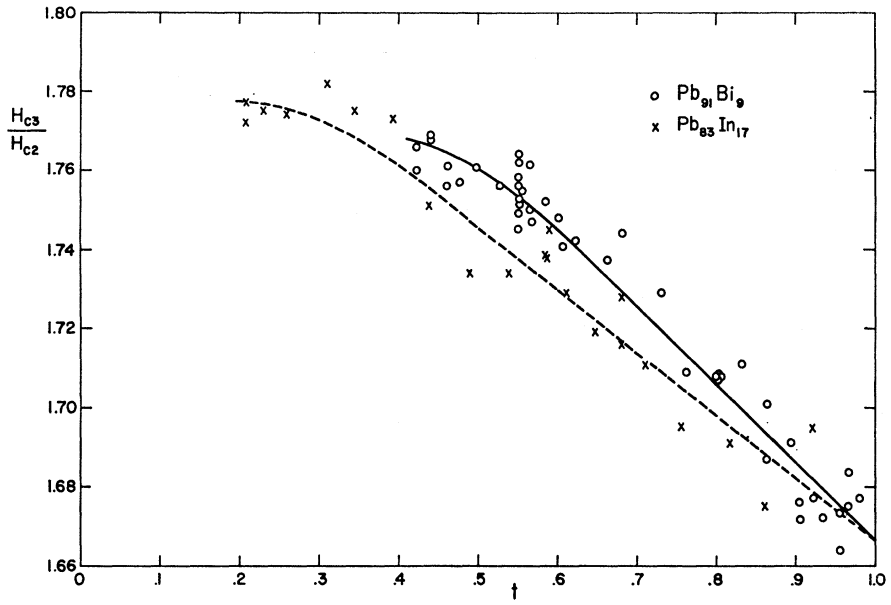

FIG. 3. Ratio H_{c3}/H_{c2} versus temperature for the two alloys $Pb_{0.91}Bi_{0.09}$ and $Pb_{0.83}In_{0.17}$.

Bi destroys the ductility of pure Pb, we are tempted to conclude that the addition to Pb of a quinquevalent element increases $N(0)$ quite markedly. From these purely metallurgical properties, then, we would conclude that in Pb the Fermi energy E_F is slightly below a density-of-states peak, contrary to what is suggested by the sketch of $N(E)$ in the work of Anderson and Gold.[20] In fact, our surface-resistance data, as we shall see in Sec. IV, give very strong evidence that E_F lies below a density-of-states peak.

The $R(H)$ measurements were carried out with two alloys, a $Pb_{0.83}In_{0.17}$ and a $Pb_{0.91}Bi_{0.09}$. Because the Bi alloy was prepared by evaporation, its composition is not very accurately known. Its thickness is about 5 μ. The rolled Pb–In sample is about 400 μ thick. During sample preparation, test strips were prepared simultaneously for the measurement of thickness and dc conductivity needed to evaluate the classical skin depth δ_0, as reported in Table I.

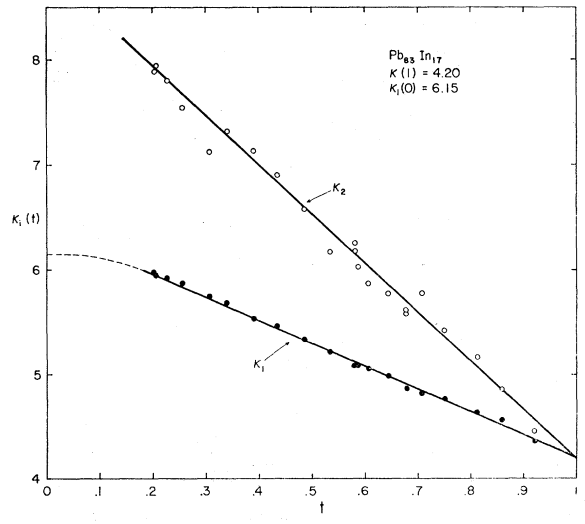

FIG. 4. $\kappa_1(t)$ and $\kappa_2(t)$ for the $Pb_{0.83}In_{0.17}$ alloy. $\kappa_1(t)$ in this figure is calculated with $h_c(t)$ of pure Pb, which is unjustified as explained in text.

[20] J. R. Anderson and A. V. Gold, Phys. Rev. **139**, A1459 (1965).

IV. ANALYSIS OF EXPERIMENTAL RESULTS

Figure 1 is a recording of $R(H)$ for the $Pb_{0.91}Bi_{0.09}$ alloy at 4.19°K. From such recordings taken at various temperatures it is easy to derive H_{c3}, H_{c2}, s_3, s_2, and r as defined in Sec. II. In Fig. 2 we see the experimental functions $s_3(t)$ and $s_2(t)$, and in Fig. 3 the ratio H_{c3}/H_{c2} found for the two alloys, which show essentially the same behavior. By plotting $H_{c2}(t)$ also against the

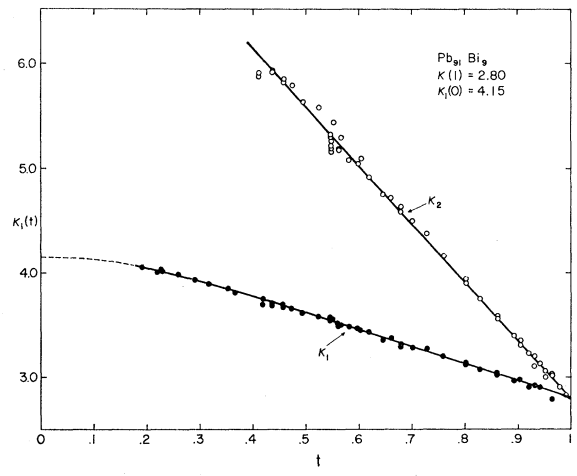

FIG. 5. $\kappa_1(t)$ and $\kappa_2(t)$ for $Pb_{0.91}Bi_{0.09}$ alloy. $\kappa_1(t)$ in this figure is calculated with $h_c(t)$ of pure Pb, which is unjustified as explained in text.

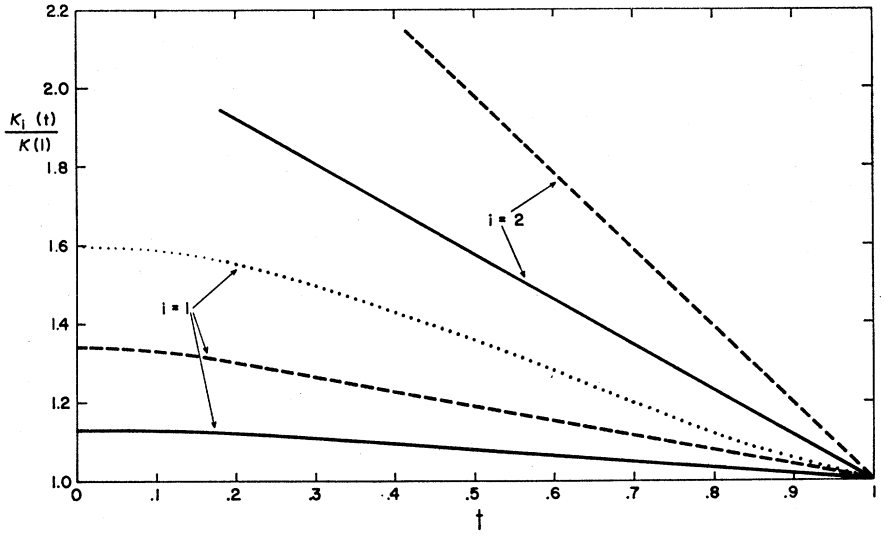

FIG. 6. $\kappa_1(t)/\kappa(1)$ and $\kappa_2(t)/\kappa(1)$ for the alloys $Pb_{0.83}In_{0.17}$ (solid lines) and $Pb_{0.91}Bi_{0.09}$ (dashed lines), and $\kappa_1(t)/\kappa(1)$ for pure Pb (Ref. 27; dotted line). For the alloys, $\kappa_1(t)/\kappa(1)$ is a mere sketch, as only the values at $t=0$ and $t=1$ have been calculated, as outlined in the text.

function $f=(1-t^2)/(1+t^2)$, one can easily find dH_{c2}/dt at $t=1$ and, by linear extrapolation toward $f=1$, the field $H_{c2}(0)$. For the In and Bi alloys we find, respectively, 5090 and 4870 Oe for $H_{c2}(0)$ and for the slopes at $t=1$ we obtain, within experimental accuracy, the value predicted by theory in the weak-coupling dirty limit,[3,14] i.e., in reduced units $-h_{c2}'(1)=1.444$. The temperature dependence of $H_{c2}(t)$ is quite close to this theory over the entire range; for both alloys the experimental points lie above the theoretical curve, with maximum deviations near $t=0.4$ of about 1.8% and 3.2% for the In and Bi alloys, respectively. For both alloys $H_{c2}(t)$ is more linear than predicted by theory near $t=1$.

If we want to derive the parameters $\kappa_1(t)$ from the measured $H_{c2}(t)$, we have to know the thermodynamic critical fields $H_c(t)$ of these alloys. But there simply are no such data available, and magnetization, which in principle can furnish such data, cannot give this data reliably because of hysteretic effects. We have, therefore, chosen a different approach. Equation (1) does not involve $H_c(t)$, and since all variables of this equation, excepting the unknown $\kappa_2(t)$, are easily accessible experimentally, we derive first $\kappa_2(t)$ as shown in Figs. 4 and 5. Once $\kappa_2(t)$ is known, one also knows $\kappa(1)=\kappa_1(1)=\kappa_2(1)=\kappa_3(1)$.[3] Since the temperature dependence of $H_{c2}(t)$ is known, all that is needed to find $\kappa_1(t)$ is the temperature dependence of the thermodynamic critical field of the alloy, i.e., in reduced units the function $h_c(t)$. If we take for $h_c(t)$ the function found experimentally[21] for pure Pb, we end up with the $\kappa_1(t)$ values of Figs. 4 and 5. As we shall see shortly, however, $\kappa_1(t)$ values obtained in this fashion are probably false.

There is an accumulation of evidence[22,23] linking coupling strength $N(0)V$ with deviations from BCS predictions[24] of the slope at T_c of the thermodynamic critical field, i.e., $-h_c'(1)$ and Δ_{00}/k_BT_c, where Δ_{00} is the energy gap at $T=0$ and $H=0$. Whereas BCS predict[24] $-h_c'(1)=1.736$ and $\Delta_{00}/k_BT_c=1.764$, one finds for Pb[18,21] $-h_c'(1)=2.13$ and $\Delta_{00}/k_BT_c=2.17$. There have been suggestions that these two parameters should always remain nearly equal and deviate upward as the coupling strength increases.[22] But there have also been arguments according to which these deviations can be somewhat more complex.[23] Nevertheless, one might say that choosing for our alloys the $h_c(t)$ function of pure Pb is equivalent to assuming our alloys to have the same coupling strength as Pb. While the Δ_{00}/k_BT_c values of Adler *et al.*[18] support this assumption, our own data, on the contrary, indicate that the coupling strength of the $Pb_{0.83}In_{0.17}$ alloy is appreciably less than that of the $Pb_{0.91}Bi_{0.09}$ alloy, which is itself less than that of pure Pb.

Three independent pieces of evidence point toward a reduced coupling strength for the Pb–In alloy:

(a) The reduced parameters $\kappa_2(t)/\kappa(1)$ for $Pb_{0.83}In_{0.17}$ and $Pb_{0.91}Bi_{0.09}$ shown in Fig. 6 exhibit a large difference. For the Bi alloy the data of Fig. 6 indicate $\kappa_2(0)/\kappa(1)\cong 3$, whereas for the In alloy the same ratio only equals about 2.

(b) When we derive $\kappa_1(t)$ from our $H_{c2}(t)$ with a particular choice of the $h_c(t)$ function, we also make a choice of $H_0=H_c(0)$, for we have

$$\kappa(1)=\frac{1}{\sqrt{2}}\frac{H_{c2}(0)}{H_0}\frac{h_{c2}'(1)}{h_c'(1)}. \tag{5}$$

If we take the $h_c(t)$ of pure Pb, we need, to satisfy Eq. (5), H_0 values of 580 Oe for $Pb_{0.83}In_{0.17}$ and 830 Oe for $Pb_{0.91}Bi_{0.09}$. Clearly, this value of H_0 for the In alloy is much too small to be acceptable. Equation (5)

[21] D. L. Decker, D. E. Mapother, and R. W. Shaw, Phys. Rev. 112, 1888 (1958).
[22] A. M. Toxen, Phys. Rev. Letters 15, 462 (1965).
[23] J. Grunzweig-Genossar and M. Revzen, Phys. Rev. Letters 16, 131 (1966).
[24] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).

suggests that H_0 would increase if $-h_c'(1)$ were to decrease.

(c) If we combine an equation from the microscopic theory,[3]

$$H_{c2}(0)=1.764(\phi_0 k_B T_c/hD), \tag{6}$$

where $\phi_0=hc/2e$ is the flux quantum, with the conductivity formula

$$\sigma=2N(0)e^2D, \tag{7}$$

we find

$$N(0)=\sigma H_{c2}(0)/1.764cek_BT_c. \tag{8}$$

For our two alloys, we derive from Eq. (8) that

$$N(0)\,|_{Pb_{0.91}Bi_{0.09}}:N(0)\,|_{Pb_{0.83}In_{0.17}}=1.25. \tag{9}$$

Even if we allow for some uncertainty as to the applicability of the weak-coupling microscopic formalism to our alloys, there is no doubt that $N(0)$ is larger for the Bi alloy than for the In alloy, and the same is certainly true for the coupling constant $N(0)V$ as well.

Equation (8) can be combined with another microscopic result,[24]

$$H_0=1.764[4\pi N(0)]^{1/2}k_BT_c, \tag{10}$$

to yield a formula for H_0:

$$H_0=\{1.764[4\pi\sigma H_{c2}(0)k_BT_c/ce]\}^{1/2}. \tag{11}$$

From this last equation we find for $Pb_{0.83}In_{0.17}$ a field H_0 of 759 Oe and for $Pb_{0.91}Bi_{0.09}$ an H_0 of 915 Oe. With these values Eq. (5) then requires $-h_c'(1)=1.63$ for the In alloy and 1.94 for the Bi alloy. These figures, which are collected in the table, are much more acceptable than the low H_0 found before. Some comment should be made, however, about the rather low values of $-h_c'(1)$ that we find and the large ratios Δ_{00}/k_BT_c recorded by Adler *et al.*[18] We think that the large difference found for $Pb_{0.83}In_{0.17}$ can be explained only in part by the argument of Grunzweig-Genossar and Revzen.[23] We also believe that Adler *et al.*,[18] who did not anneal their alloys, had inhomogeneous material. Our own observations clearly showed that for both types of alloys, Pb–In and Pb–Bi, homogenous material was not obtained unless it was annealed for at least 24 h at about 50°C below its melting point. The inhomogeneity does not manifest itself very much in the resistivity measurements but shows very strongly in the $R(H)$ and critical-field data. Homogenous alloys give strongly featured $R(H)$ curves, as shown in Fig. 1, and the "observed" H_{c2} and H_{c3} fields become more and more clearly defined, and drop by large amounts, as the alloys become more and more homogenous when the annealing time is successively increased. Eventually, they reach a stable low value typical of the homogenous alloy.

V. DISCUSSION AND CONCLUSIONS

We have been able to calculate the microwave surface impedance of dirty type-II superconductors, in the surface-sheath regime, in terms of the microscopic theory of superconductivity.[3] Experimenting with two lead alloys, we have then demonstrated the consistency of our theory and the suitability of surface-impedance measurements near H_{c3} to derive the parameters that characterize the properties of a superconductor. The theory is strictly valid in the weak-coupling limit, but our experimental results suggest that strong-coupling effects do not impair the applicability of weak-coupling formalism, but produce only deviations for $H_c(t)$, $H_{c2}(t)$, and $H_{c3}(t)$; these can be accounted for by stronger variations with temperature than predicted by weak-coupling theories[25] of the two Ginzburg-Landau–type parameters $\kappa_1(t)$ and $\kappa_2(t)$ defined by Maki.[3] These findings give support to the strong-coupling calculation of Werthamer and McMillan[26] and to recently published $H_{c2}(t)$ data for pure Pb.[27] Figure 3 also suggests that the ratio $H_{c3}(t)/H_{c2}(t)$ deviates by less than 5% over the entire range $0\leq t\leq 1$ from the theoretical value at T_c of 1.695. The possibility cannot be entirely excluded that this variation is not characteristic of a homogeneous superconductor, but is indicative of an increase of κ toward the surface. Such an increase would be rather ineffective at T_c because of the larger coherence length $\xi(t)$ as $t\to 1$. However, since the function $\xi(t)/\xi(0)$ increases only from 1 to 2 as t goes from 0 to 0.83, one would have expected the ratio of $H_{c3}(t)/H_{c2}(t)$ to vary much more rapidly just below $t=1$ and level off very soon to its low-temperature value, unlike the rather gradual behavior seen in Fig. 3.

The $\kappa_2(t)$ functions of Figs. 4 and 5 are strikingly linear. The $\kappa_1(t)$ data reported in these figures are obtained by choosing a thermodynamical field $H_c(t)$ that has the same temperature dependence as $H_c(t)$ of pure Pb. As we have seen, this implies unacceptable values of H_0. The weak-coupling formalism leads to the H_0 and $\kappa_1(0)$ values of the table, and from these we find $\kappa_1(0)/\kappa(1)=1.34$ and 1.13 for the $Pb_{0.91}Bi_{0.09}$ and $Pb_{0.83}In_{0.17}$ alloys, respectively. The last of these values is unexpectedly low. It may bear testimony to the restricted applicability of the weak-coupling formalism, or mean that the effective transport mean free path l_{tr} is larger than the s-scattering-limited electron mean free path[25] l. In any case, it suggests again that $Pb_{0.83}In_{0.17}$ is in fact a rather weak-coupling superconductor, even though its $\kappa_2(t)$ variation is somewhat stronger than predicted by weak-coupling theory.[25]

The variations of $\kappa_2(t)$ reported here are far larger than those quoted by Bon Mardion *et al.*,[28] whose magnetization measurements on Pb–Tl alloys yielded $\kappa_2(t)$ in a rather indirect way, so that not much significance should be attached to this discrepancy.

The sheath thickness, characterized by the parameter $\alpha(t)$ plotted in Fig. 7, shows a systematic behavior as

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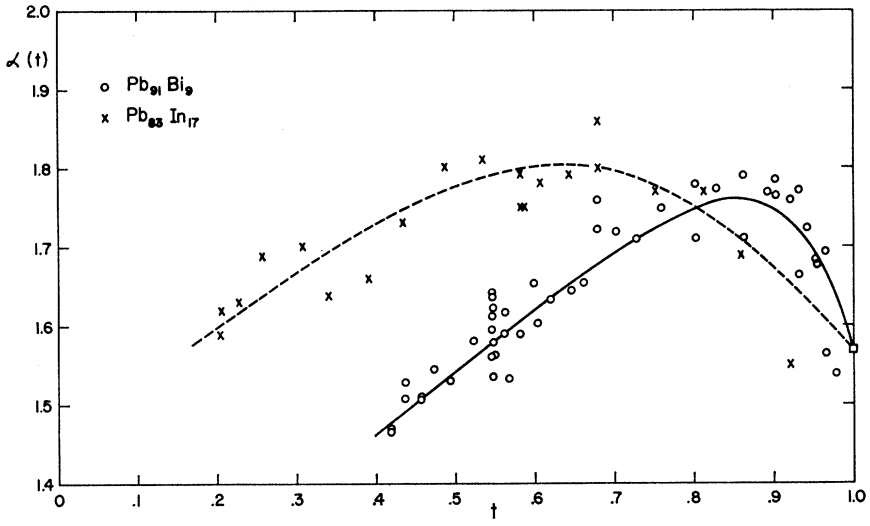

FIG. 7. Sheath-thickness parameter $\alpha(t)$ defined by Eq. (4). The square at $t=1$ is from theory (see Ref. 10).

function of temperature. At T_c we find agreement with the Ginzburg-Landau limit calculated by Fink and Kessinger.[10] There are obvious similarities and differences for the two alloys in the behavior of $\alpha(t)$ below T_c. We have no explanation to offer accounting for the observed behavior, but must point out that it may again be connected with an increase of κ toward the surface. If this were so, $\alpha(t)$ would be expected to rise as t decreased from $t=1$. For a superconductor perfectly homogeneous right up to the surface, one might have expected $\alpha(t)$ to be monotonic with t. The behavior observed and the perturbation envisaged as possible then indicate that the unperturbed $\alpha(t)$ decreases monotonically with t. As discussed before, it is also quite likely that α is not subject to any perturbation and that the $\alpha(t)$ observed is genuine.

APPENDIX: SURFACE IMPEDANCE OF INHOMOGENEOUS SUPERCONDUCTORS

We assume that the superconductor occupies the halfspace $x<0$ and that the dc magnetic field $\mathbf{H}$ is parallel to the surface, pointed in the z direction.[29] The monochromatic plane-polarized microwave is incident perpendicularly to the surface, with its electric vector $\mathbf{E}_\omega$ parallel to $\mathbf{H}$. In this situation the microscopic calculation gives, for the current induced by the microwave vector potential $\mathbf{A}_\omega$, the following expression[11]:

$$\mathbf{j}_\omega(\mathbf{r})=Q(\mathbf{r},\omega)\mathbf{A}_\omega(\mathbf{r}), \tag{A1}$$

with

$$Q(\mathbf{r},\omega)=\sigma\Big\{-i\omega-\frac{|\Delta(\mathbf{r})|^2}{2\pi T}\Big[\psi^{(1)}(\tfrac{1}{2}-i\omega/2\pi T+\rho)+\Big(\frac{2\pi T}{-i\omega}+\frac{2\pi T}{-i\omega+\epsilon_0}\Big)[\psi(\tfrac{1}{2}-i\omega/2\pi T+\rho)-\psi(\tfrac{1}{2}+\rho)]\Big]\Big\}, \tag{A2}$$

where σ is the normal-state conductivity (assumed entirely real),

$$\rho=\epsilon_0/4\pi T, \qquad \epsilon_0=2DeH_{c2}(t), \qquad D=\tfrac{1}{3}v_F l$$

is the diffusion constant, and ψ and $\psi^{(1)}$ are the di- and trigamma functions. $\Delta(\mathbf{r})$ is the spatially variable order parameter.

The expression (A2) is valid in the surface-sheath regime near H_{c3} with a $\Delta(\mathbf{r})$ as first calculated by Saint-James and de Gennes,[1] or in the mixed state near H_{c2} where $\Delta(\mathbf{r})$ is given by the Abrikosov solution.[11,30] It is important to stress that (A2) holds only for the geometry considered, $\mathbf{E}_\omega \parallel \mathbf{H}$, where we can completely neglect contributions from fluctuations of the order parameter.[5]

In order to calculate the surface impedance, we have to solve the differential equation

$$-\partial^2 A_\omega(x)/\partial x^2=4\pi Q(x,\omega)A_\omega(x) \tag{A3}$$

with appropriate boundary conditions. The surface impedance is then given by

$$Z=R+iX=4\pi\frac{E_\omega(0)}{H_\omega(0)}=-4\pi i\omega\left.\frac{A_\omega(x)}{(\partial/\partial x)A_\omega(x)}\right|_{x=0}. \tag{A4}$$

We consider now the two regimes where (A2) applies.

1. Surface Sheath, $H \lesssim H_{c3}$

Since for $x\gg\xi(t)$, i.e., far inside the superconductor, $Q(x,\omega)$ is a constant equal to its normal-state value, we can write (A3) in the form

$$-\partial^2 A_\omega(x)/\partial x^2=-4\pi\sigma[i\omega+f(x)]A_\omega(x), \tag{A5}$$

where $f(x)\propto|\Delta(x)|^2$ is a small perturbation, because we are close to H_{c3}. Since the vector potential decreases inside the superconductor in a manner only slightly different from the way it does in the normal state, we

[29] The calculation presented in the Appendix is carried out in a system of units such that $\hbar=k_B=c=1$, and with a time dependence of the form $\exp(+i\omega t)$.

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look for solutions of (A5) of the form

$$A_\omega(x)=A_\omega(0)\ \exp[-bx-\varphi(x)], \tag{A6}$$

where $\varphi'(\infty)=0$, $|\varphi(x)|\ll 1$, and

$$b^{-1}=(4\pi i\omega\sigma)^{1/2}=(1-i)\delta_n=\delta_0/(1+i), \tag{A7}$$

with

$$\delta_0=2\delta_n=(2\pi\omega\sigma)^{-1/2}.$$

δ_0 is called the normal-state classical skin depth.

Substituting (A6) in (A5), we obtain

$$2b\varphi'(x)+[\varphi'(x)]^2-\varphi''(x)=4\pi\sigma f(x). \tag{A8}$$

Neglecting $[\varphi'(x)]^2$ in (A8), we have

$$\varphi(x)=4\pi\sigma\int_0^x dx\left[e^{2bx}\int_x^\infty e^{-2bx'}f(x')\,dx'\right]. \tag{A9}$$

The surface impedance is then given by

$$Z=4\pi i\omega/[b+\varphi'(0)], \tag{A10}$$

where

$$\varphi'(0)=4\pi\sigma\int_0^\infty e^{-2bx'}f(x')\,dx'. \tag{A11}$$

In the following we consider only the low-frequency behavior of Z and assume that $\delta_0\gg\xi(t)$. Then $f(x)$ is given by

$$f(x)=\{|\Delta(x)|^2/\pi T\}\psi^{(1)}(\tfrac{1}{2}+\rho). \tag{A12}$$

If we approximate $\Delta(x)$ with the Gaussian form

$$\Delta(x)=\Delta(0)\ \exp[-x^2/2\xi^2(t)], \tag{A13}$$

we can use a relation derived by Maki[11] for $\langle|\Delta(x)|^2\rangle_{Av}$ normalized in the sheath thickness $\xi(t)$,

$$\langle|\Delta(x)|^2\rangle_{Av}=(\tfrac{1}{2}\pi)^{1/2}\frac{0.59eT}{\sigma}\frac{H_{c3}-H}{2\kappa_2^2(t)-0.334}(\psi^{(1)})^{-1}, \tag{A14}$$

and we can calculate

$$\varphi'(0)=4\pi\sigma\int_0^\infty f(x)\,dx$$

$$=\frac{(2\pi)^{1/2}}{\xi(t)}\left(\frac{H_{c3}-H}{H_{c3}}\right)[2\kappa_2^2(t)-0.334]^{-1}. \tag{A15}$$

For the surface impedance we find finally

$$Z=\frac{4\pi i\omega}{(1+i)\delta_0^{-1}+(2\pi)^{1/2}\xi^{-1}(t)[(H_{c3}-H)/H_{c3}][2\kappa_2^2(t)-0.334]^{-1}} \tag{A16}$$

or

$$Z\cong R_n\left[1+i-(2\pi)^{1/2}\frac{\delta_0}{\xi(t)}\left(\frac{H_{c3}-H}{H_{c3}}\right)[2\kappa_2^2(t)-0.334]^{-1}\right], \tag{A17}$$

and we can derive

$$s_3(t)=(H_{c3}/R_n)[\partial R(H)/\partial H]|_{H=Hc3}=(2\pi)^{1/2}[\delta_0/\xi(t)][2\kappa_2^2(t)-0.334]^{-1}. \tag{A18}$$

2. Transition into the Mixed State, $H\cong H_{c2}$

We consider now the discontinuous jump of $\partial R(H)/\partial H$ at $H=H_{c2}$. We can use again (A10) and (A11) to calculate Z, although we have no explicit expressions with which to evaluate the integral in (A11). We shall assume that the linear relationship with H obtained for this integral can still be used, with a factor $\alpha(H,t)$ which expresses the departure from linearity. $\alpha(H,t)$ is generally >1 and can be looked upon as the ratio of sheath thicknesses at the fields H and H_{c3}. Fink and Kessinger[10] have calculated $\alpha(H,t)$ with the help of the Ginzburg-Landau equations. Since these equations are valid near T_c, one would expect that their result for $\alpha(H,t)$ is valid near T_c, as our experiments confirm. We thus write for (A15)

$$\varphi'(0)=4\pi\sigma\int_0^\infty f(x)\,dx$$

$$=\frac{(2\pi)^{1/2}\alpha(H,t)}{\xi(t)}\left(\frac{H_{c3}-H}{H_{c3}}\right)[2\kappa_2^2(t)-0.334]^{-1}. \tag{A19}$$

Then we express $R(H_{c2})/R_n$ in terms of $\alpha(t)=\alpha(H_{c2},t)$:

$$r=R(H_{c2})/R_n=[(1+A)^2+A^2]^{-1}, \tag{A20}$$

where

$$A=(\tfrac{1}{2}\pi)^{1/2}\frac{\delta_0\alpha}{\xi(t)}\left(\frac{H_{c3}-H_{c2}}{H_{c3}}\right)[2\kappa_2^2(t)-0.334]^{-1}. \tag{A21}$$

In fields below H_{c2} one has the Abrikosov phase in the bulk. We can carry out all the calculations in a way similar to that of (1) above, and we obtain,

$$Z=4\pi i\omega\Big/\left[b\left(1+\frac{ie}{2\pi\omega\sigma}\frac{H_{c2}-H}{[2\kappa_2^2(t)-1]\beta_A}\right)+\varphi'(0)\right], \tag{A22}$$

where $\beta_A=1.16$, b is the same as before [Eq. (A7)], and $\varphi'(0)$ is still given by (A19). The coefficient of b in the denominator describes the effect of the Abrikosov structure[11,30] on the surface impedance. One can then

calculate

$$s_2(t)=\frac{H_{c2}}{R_n}\left(\frac{\partial R(H)}{\partial H}\right)\Bigg|_{H=H_{c2}-0}-\frac{H_{c2}}{R_n}\left(\frac{\partial R(H)}{\partial H}\right)\Bigg|_{H=H_{c2}+0}$$

$$=\frac{g(r)}{2\beta_A}\left(\frac{\delta_0}{\xi(t)}\right)^2[2\kappa_2^2(t)-1]^{-1}, \quad \text{(A23)}$$

with

$$g(r)=r[1-r+r(2/r-1)^{1/2}].$$

Finally, by combining (A18), (A20), and (A21), we can derive

$$\alpha(t)=\frac{(2/r-1)^{1/2}-1}{s_3(t)}\left(\frac{H_{c3}}{H_{c3}-H_{c2}}\right). \quad \text{(A24)}$$

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Electron Correlations at Metallic Densities*

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The dielectric function of a degenerate electron gas in the random-phase approximation, and the one proposed by Hubbard, which takes exchange effects into account, have been extensively used in the study of metallic properties. However, both dielectric functions lead to an overestimate of the short-range correlations between particles. This is manifest from the fact that the pair-correlation function is negative for small interparticle separations over the whole range of metallic densities, and implies an overestimate of the correlation energy. An improved expression of the dielectric function is given, which includes explicitly, in an approximate way, the short-range correlations arising from both Coulomb and exchange effects by being a functional of the structure factor. The structure factor and the dielectric function can then be determined in a self-consistent manner. The numerical solution of the self-consistent scheme yields a pair-correlation function which is positive for all values of the density up to $r_s=4$. For $r_s>4$, it is very slightly negative at small separations, but it is so small that it can be considered to be zero for all practical purposes. New estimates of the correlation energy are given for the entire metallic density range, and are smaller than the earlier estimates. These results are used to recalculate the cohesive energy of the alkali metals. A discussion of the plasmon dispersion relation, the compressibility, and the liquid-solid transition, both for the electron system and for an astrophysically interesting system of protons over a background of electrons, is also given.

I. INTRODUCTION

THE dielectric formulation of the many-body problem has been found to be very fruitful in studying the degenerate electron gas and the metallic properties which depend strongly on electron-electron interactions. The system that one studies is a degenerate electron gas on a uniform, neutralizing background. The density-fluctuation excitation spectrum, the correlations between the density fluctuations, and the ground-state energy of the system are rigorously expressible in terms of its frequency- and wavelength-dependent dielectric function. This model system serves as a useful guide to the study of many metallic properties, such as the interionic potential and the screening of defects, under the assumption that the dielectric function is not essentially altered by the discrete nature of the ion lattice. It is, therefore, of great importance to have a precise knowledge of this function in the range of electron densities encountered in metals.

The dielectric function first given by Lindhard,[1] which corresponds to the random-phase approximation[2–4] (RPA), is the one which is most commonly used. It provides a good description of the plasmon excitation modes and of long-wavelength screening phenomena, but its validity is otherwise limited to high electron densities ($r_s \ll 1$). The inadequacy of RPA becomes manifest, for instance, from the fact that the pair-distribution function, which is positive-definite, becomes negative[5,6] for small separation between particles, over the entire range of metallic densities ($2 \leq r_s \leq 6$). This arises from the failure of the RPA to take account of short-range effects; indeed, no local field correction[7] is made in this theory. The neglect of short-range

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